Polypropylene-based resin composition, process for producing the same and injection molded article

BACKGROUND OF THE INVENTION

5 Field of the Invention

The present invention relates to a polypropylene-based resin composition, a process for producing the same and an injection molded article made of the same. More particularly, the present invention relates to a polypropylene-based resin

10 composition which, when made into a molded article, has a low tendency of generation of flow marks, namely, a high die swell, and excellent appearance such as little generation of granular structures(fish eye), and is excellent in balance of rigidity and toughness, a process for producing the same and an injection

15 molded article made of the same.

Description of Related Arts

Polypropylene-based resin compositions are materials excellent in rigidity, impact resistance and the like, and

20 utilized in wide applications as molded article such as automobile interior and exterior materials, electric appliance housings and the like. It is conventionally well known that, among these polypropylene-based resin compositions, polypropylene-based resin compositions containing a

25 propylene-ethylene block copolymer, for example, polypropylene-based resin compositions composed of a propylene-ethylene block copolymer and a propylene homopolymer,

or of different two or more propylene-ethylene block copolymers, are excellent in rigidity, impact resistance and the like and suitably used.

For example, JP07-157626A discloses a thermoplastic resin composition containing a propylene-ethylene block copolymer composed of a propylene homopolymer phase and a propylene-ethylene copolymer phase obtained by multi-stage polymerization and polyolefin rubber. The propylene-ethylene block copolymer is composed of a propylene-ethylene random copolymer phase(i) having an ethylene content of from 5 to 50% by weight and an intrinsic viscosity of from 4.0 to 8.0 dl/g, and a propylene-ethylene block copolymer phase (ii) having an ethylene content of over 50% by weight and 98% by weight or less and an intrinsic viscosity of 2.0 dl/g or more and less than 4.0 dl/g, and it is disclosed that a thermoplastic resin composition having an extremely high ductility is obtained.

JP07-157627A discloses a thermoplastic resin composition containing a propylene-ethylene block copolymer obtained by multi-stage polymerization and polyolefin-based rubber. The propylene-ethylene block copolymer is composed of a propylene-ethylene block copolymer in which the propylene-ethylene random copolymer phase(iii) has an intrinsic viscosity of from 4.0 to 8.0 dl/g, and a propylene-ethylene block copolymer in which the propylene-ethylene block copolymer phase (iv) has an intrinsic viscosity of 2.0 dl/g or more and less than 4.0 dl/g, (excluding a propylene-ethylene block copolymer having a

propylene-ethylene random copolymer phase having an intrinsic viscosity of from 4.0 to 8.0 dl/g and an ethylene content of from 5 to 50% by weight and a propylene-ethylene block copolymer having a propylene-ethylene random copolymer phase having an

5   intrinsic viscosity of 2.0 dl/g or more and less than 4.0 dl/g and an ethylene content of over 50% by weight and 98% by weight or less), and it is disclosed that the thermoplastic resin composition having extremely high ductility is obtained.

JP07-233305A discloses a polypropylene resin composition
10 having improved melt index, composed of a polypropylene, a inorganic filler and a metal soap of zinc salt of a fatty acid having 15 to 20 carbon atoms. As the polypropylene, block copolymers containing a propylene-ethylene copolymer part having a [η] of 2 to 6 dl/g and a propylene-ethylene copolymer
15 part having a [η] of 7 to 15 dl/g are described, and as a preferable embodiment, those composed of at least two kinds of block copolymers are described. Further, it is described that a polypropylene resin composition manifesting no practically problematical lowering in impact strength, rigidity, heat
20 resistance and the like, having improved flowability and suitable for producing a molded article thinned in thickness, and showing little discoloration to yellow and pink caused by pelletizing and little bleeding of a low molecular compound on the surface of a mold and the surface of an injection molded
25 article, is obtained.

In addition, a propylene-ethylene block copolymer suitably used due to excellent rigidity, impact resistance and

the like is recently produced by a continuous gas phase polymerization method because of simple production process and production at low cost. However, the propylene-ethylene block copolymer produced by the gas phase method generally has problems

5   of poor appearance of a molded article due to generation of granular structures when the intrinsic viscosity of a propylene-ethylene random copolymer portion is set at high level.

For solving such problems on appearance, for example,

10 JP07-286075A discloses a propylene polymer composition composed of a propylene polymer produced by a continuous polymerization method in which the 23℃ n-decane-soluble component content is 0% by weight or more and less than 15% by weight and the 23℃ n-decane-soluble component contains a

15 structural unit derived from ethylene in an amount of 30 to 60 mol% and has an intrinsic viscosity [η] of 3 to 7 dl/g, and a propylene block copolymer produced by a batch-wise solvent polymerization method or continuous solvent polymerization method in which the 23℃ n-decane-soluble component content

20 is 15 to 40% by weight and this 23℃ n-decane-soluble component contains a structural unit derived from ethylene in an amount of 30 to 60 mol% and has an intrinsic viscosity [η] of 5 to 12 dl/g, and it is described that a propylene polymer composition having excellent rigidity and impact resistance and capable

25 of forming a molded article without generating granular structures on appearance is obtained, however, there is desired a further improvement regarding generation of granular

structures.

On the other hand, WO98/54233 discloses a polypropylene resin composition in which the content of a high molecular weight polypropylene having an intrinsic viscosity [η] of 9 to 13

5   dl/g is from 15 to 30% by weight, the molecular weight distribution represented by Mw/Mn is 20 or more, the molecular weight distribution represented by Mz/Mw is 7 or more, and when molded into an injection molded article, the thickness of a skin layer formed on the surface of this injection molded article

10   is 300 μm or more, and it is described that a polypropylene resin composition which has light weight, has extremely high rigidity and heat resistance and manifests excellent flowability in molding and excellent in processability can be produced.

15   However, it is described in, for example, Plastics Age, May, 1980, p. 93, Fig. 2. 27 that when the thickness of a skin layer formed on the surface of an injection molded article increases, ultimate elongation (tensile elongation), namely, toughness lowers, and there is a desire for an improvement of

20   balance between rigidity and toughness regarding the above-mentioned polypropylene resin composition described in WO98/54233.

JP2000-226478A describes a polypropylene resin composition having a molecular weight distribution represented

25   by Mw/Mn of 8 or more and containing a high molecular weight polypropylene having an intrinsic viscosity [η] of 6 to 11 dl/g and a low molecular weight polypropylene having an intrinsic

viscosity $[\eta]$ of 0.6 to 1.6 dl/g, and it is described that a polypropylene resin composition which has light weight, has extremely high rigidity and heat resistance and, shows excellent flowability in molding, generates no cracking in molding and

5 excellent in moldability can be provided. However, since the ultimate elongation of the above-mentioned polypropylene resin composition described in an example of JP-A No. 2000-226478 is from 8 to 16%, there is desired an improvement in balance between rigidity and toughness.

10 As described above, there is a desire for the development of a polypropylene-based resin composition which, when molded into a molded article, manifests a low tendency of generation of flow mark on the surface of the molded article, namely, has excellent appearance properties such as high die swell and

15 practically no-problematical generation of granular structures, and is excellent in balance between rigidity and toughness.

SUMMARY OF THE INVENTION

An object of the present invention is to provide a

20 polypropylene-based resin composition which, when molded into a molded article, manifests a low tendency of generation of flow marks, namely has a high die swell, has excellent appearance such as little generation of granular structures, and the like, and is excellent in balance between rigidity and toughness,

25 to a process for producing the same, and to an injection molded article made of the same.

Namely, the present invention relates to a

polypropylene-based resin composition comprising 0.5 to 10% by weight of a propylene-based polymer component (A1) obtained by polymerizing a monomer mainly composed of propylene, having an intrinsic viscosity $[\eta]^{A1}$ measured in tetralin at 135°C of

5   5 dl/g or more and a melting peak temperature Tm^{A1} measured by a differential scanning calorimeter(herein-after, referred to as "DSC".) of 130 to 160°C, and 90 to 99.5% by weight of a propylene-based polymer component (A2) obtained by polymerizing a monomer mainly composed of propylene, having

10   an intrinsic viscosity $[\eta]^{A2}$ measured in tetralin at 135°C of less than 5 dl/g, a process for producing the same and an injection molded article made of the same.

The present invention will be described in detail below.

15   **DETAILED DESCRIPTION OF THE INVENTION**

The propylene-based polymer component (A1) used in the present invention is a propylene-based polymer obtained by polymerizing a monomer mainly composed of propylene, and examples thereof include a propylene homopolymer obtained by

20   homo-polymerization of propylene, a propylene-ethylene random copolymer obtained by copolymerization of propylene and ethylene, a propylene-α-olefin random copolymer obtained by copolymerization of propylene and an α-olefin having 4 to 12 carbon atoms, and the like, and preferable is a

25   propylene-ethylene random copolymer.

The ethylene content in the propylene-ethylene random copolymer is usually from 0.5 to 8% by weight, preferably from

1 to 7% by weight.

As the α-olefin having 4 to 12 carbon atoms used in a propylene-α-olefin random copolymer, butene-1, pentene-1, hexene-1, 4-methylpentene-1, octane-1, decene-1 and the like

5 are listed, and preferable is butene-1.

The content of the α-olefin is usually from 1 to 20% by weight, preferably from 2 to 15% by weight.

As the propylene-α-olefin random copolymer, for example, a propylene-butene-1 random copolymer, propylene-hexene-1

10 random copolymer and the like are listed, and preferable is a propylene-butene-1 random copolymer.

The intrinsic viscosity $[\eta]^{A1}$ measured in tetralin at 135℃ of the propylene-based polymer component (A1) used in the present invention is 5 dl/g or more, preferably 6 dl/g or

15 more, and from the standpoint of dispersibility of the component (A1), further preferably 6 to 9 dl/g. When the intrinsic viscosity $[\eta]^{A1}$ is less than 5 dl/g, high die swell is not obtained, and prevention of generation of flow mark may be insufficient in some cases.

20 The melting peak temperature Tm^{A1} measured by a DSC of the propylene-based polymer component (A1) used in the present invention is from 130 to 160℃, preferably from 135 to 155℃. Herein, the melting peak temperature Tm^{A1} is determined from a temperature-rising thermogram of the component (A1) obtained

25 by using a DSC. When Tm^{A1} is less than 130℃, high rigidity may not be obtained, and when over 160℃, sufficient tensile elongation may not be obtained, and due to insufficient

dispersion of the component (A1), a lot of granular structure may be formed.

The propylene-based polymer component (A2) used in the present invention is a propylene-based polymer obtained by

5   polymerizing a monomer mainly composed of propylene, and specific examples thereof include a propylene homopolymer obtained by homo-polymerization of propylene, a propylene-ethylene random copolymer obtained by copolymerization of propylene and ethylene, a propylene-α

10 -olefin random copolymer obtained by copolymerization of propylene and an α-olefin having 4 to 12 carbon atoms, a propylene-ethylene block copolymer and the like, and also as the α-olefin having 4 to 12 carbon atoms, the same compounds as listed above are mentioned.

15 The intrinsic viscosity $[\eta]^{A2}$ measured in tetralin at 135℃ of the propylene-based polymer component (A2) is less than 5 dl/g, preferably 0.7 or more and less than 5 dl/g, more preferably 0.9 to 4 dl/g.

The content of the propylene-based polymer component (A1)

20 used in the present invention is from 0.5 to 10% by weight, preferably from 1 to 7% by weight. Herein, the total of the propylene-based polymer components (A1) and (A2) is 100 % by weight. Namely, the content of the propylene-based polymer component (A2) is from 90 to 99.5% by weight, preferably from

25 93 to 99% by weight. When the content of the propylene-based polymer component (A1) is less than 0.5% by weight, high die swell is not obtained, and prevention of generation of a flow

mark may be insufficient, and when the content of the propylene-based polymer component (A1) is over 10% by weight, the melt flow rate (MFR) of the polypropylene resin composition may lower and the flowability thereof may decrease.

From the standpoint of balance between rigidity and toughness of the polypropylene-based resin composition of the present invention, the propylene-based polymer component (A2) is preferably a composition (A2-1) having an intrinsic viscosity $[\eta]^B_P$ of less than 5 dl/g, composed of a propylene polymer component (B) having an intrinsic viscosity $[\eta]^B_P$ of 1.5 dl/g or less and a propylene-ethylene random copolymer component (C) having an intrinsic viscosity $[\eta]^C_{EP}$ of 8 dl/g or less and an ethylene content of 20 to 70% by weight;

a polypropylene-based resin composition(D)(A2-2) having an intrinsic viscosity $[\eta]^B_P$ of less than 5 dl/g, which is a propylene-ethylene block copolymer (i) composed of a propylene polymer component (B) having an intrinsic viscosity $[\eta]^B_P$ of 1.5 dl/g or less and a propylene-ethylene random copolymer component (C) having an intrinsic viscosity $[\eta]^C_{EP}$ of 8 dl/g or less and an ethylene content of 20 to 70% by weight, or a mixture(iii) of the propylene-ethylene block copolymer (i) and a propylene polymer (ii); or

a composition(A2-3) having an intrinsic viscosity $[\eta]^B_P$ of less than 5 dl/g, composed of a propylene polymer component (B) and the polypropylene-based resin (D).

The compositions(A2-1), (A2-2) and (A2-3) are explained in detail below.

The composition (A2-1) is composed of the propylene polymer component(B) and a propylene-ethylene random copolymer component(C). The propylene polymer component(B) is a propylene homopolymer or a propylene-α-olefin random copolymer

5 obtained by copolymerization of propylene and an α-olefin having 4 to 12 carbon atoms. As the α-olefin having 4 to 12 carbon atoms and propylene-α-olefin random copolymer, those listed above are specifically mentioned.

The intrinsic viscosity $[\eta]^B_P$ of the propylene polymer

10 component (B) is preferably 1.5 dl/g or less, further preferably from 0.7 to 1.5 dl/g from the standpoints of the melt flow rate (MFR) and flowability of the polypropylene-based resin composition.

The isotactic pentad fraction measured and determined

15 using ^{13}C-NMR of the propylene polymer component(B) used in (A2-1) is preferably 0.95 or more, further preferably 0.97 or more from the standpoints of rigidity, heat resistance and the like.

The propylene-ethylene random copolymer component (C)

20 is a propylene-ethylene random copolymer preferably having an ethylene unit content $[(C2')_{EP}]$ of 20 to 70% by weight, more preferably 25 to 65 % by weight from the standpoint of impact resistance.

The intrinsic viscosity $[\eta]^C_{EP}$ of the propylene-ethylene

25 random copolymer component (C) is preferably 8 dl/g or less, more preferably 6 dl/g or less, further preferably 1.5 to 4 dl/g, from the standpoints of prevention of significant

generation of granular structure on a molded article.

The above-mentioned granular structure is a small block mainly composed of a propylene-ethylene random copolymer component(C), generating due to poor dispersibility of the propylene-ethylene random copolymer component(C) in a polypropylene-based resin composition, and the size thereof is about 100 to several hundreds μm. In the case of formation of a molded article by injection molding or the like using a material containing a lot of granular structures, not only the appearance of the surface of the molded article is deteriorated but also bad influences are exerted on mechanical properties such as impact resistance and the like.

Preferably, the propylene polymer component(B) is a propylene homopolymer.

The compounding ratio of (A2-1) composed of the propylene polymer component (B) and propylene-ethylene random copolymer component(C) in the polypropylene-based resin composition of the present invention is from 90 to 99.5% by weight (namely, the compounding ratio of a propylene-based polymer component (A1) is from 0.5 to 10% by weight, and the total amount of (A1) and (A2-1) is 100 % by weight.), and the contents of the component(B) and the component(C) in the polypropylene-based resin composition of the present invention is preferably from 50 to 94.5% by weight and 5 to 40 % by weight(herein, the total amount of the components (A1), (B) and (C) is 100 % by weight), further preferably from 60 to 94.5% by weight and 5 to 30 % by weight, respectively.

The polypropylene-based resin composition(D)(A2-2) is a propylene-ethylene block copolymer(i), or a mixture(iii) of the propylene-ethylene block copolymer(i) and the propylene homopolymer (ii).

5   The propylene-ethylene block copolymer(i) is composed of a propylene polymer portion(first segment of the (i)) and a propylene-ethylene random copolymer portion(second segment of (i)).

The propylene polymer(ii) used in the polypropylene-based
10   resin(D) is a propylene homopolymer obtained by homo-polymerization of propylene or a propylene-α-olefin random copolymer obtained by random copolymerization of propylene and an α-olefin having 4 to 12 carbon atoms having 4 to 12 carbon atoms. As the α-olefin having 4 to 12 carbon
15   atoms and propylene-α-olefin random copolymer, those listed above are specifically mentioned.

When the polypropylene-based resin (D)(A2-2) is a mixture(iii) of the propylene-ethylene block copolymer(i) and propylene polymer(ii), the propylene polymer component(B) of
20   the polypropylene-based resin(D) means both of the propylene polymer portion which is the first segment of the propylene-ethylene block copolymer(i), and a propylene polymer(ii), and the propylene-ethylene random copolymer component(C) of the polypropylene-based resin (D) means the
25   propylene-ethylene random copolymer portion which is the second segment of the propylene-ethylene block copolymer(i).

As the propylene-ethylene block copolymer(i) and

propylene polymer(ii) used in the polypropylene-based resin (D)(A2-2), two or more propylene-ethylene block copolymers, two or more propylene polymers or mixtures thereof optionally used, respectively.

5   The intrinsic viscosity $[\eta]^B_P$ of the propylene polymer component(B) constituting the polypropylene-based resin (D)(A2-2) is preferably 1.5 dl/g or less, further preferably from 0.7 to 1.5 dl/g from the standpoints of the melt flow rate(MFR) and flowability of a polypropylene-based resin 10   composition.

The propylene polymer component(B) is preferably a propylene homopolymer from view point of rigidity and heat resistance. The isotactic pentad fraction measured and determined using ^{13}C-NMR of the propylene polymer component (B) 15   constituting the polypropylene-based resin (D)(A2-2) is preferably 0.95 or more, further preferably 0.97 or more.

The intrinsic viscosity $[\eta]^C_{EP}$ of the propylene-ethylene random copolymer component(C) used in (A2-2) constituting the polypropylene-based resin(D)(A2-2) is preferably 8 dl/g or less, 20   more preferably 6 dl/g or less, further preferably 1.5 to 4 dl/g, from the standpoints of prevention of significant generation of granular structures on a molded article.

The ethylene content $[(C2')_{EP}]$ in the propylene-ethylene random copolymer component(C) constituting the 25   polypropylene-based resin(D)(A2-2) is preferably from 20 to 70% by weight, more preferably from 25 to 65% by weight, from the standpoint of impact resistance. The balance is propylene

unit.

Regarding the a propylene polymer component (B) and the propylene-ethylene random copolymer component(C) constituting the polypropylene-based resin (D)(A2-2), the amount of a propylene-ethylene random copolymer component(C) is preferably from 5 to 40% by weight (namely, the ratio of a propylene polymer component (B) is from 95 to 60% by weight), further preferably, the ratio of a propylene-ethylene random copolymer component (C) is from 5 to 30% by weight, (namely, the ratio of a propylene polymer component (B) is from 95 to 70% by weight), wherein the total amount of a propylene polymer component (B) and a propylene-ethylene random copolymer component (C) is 100% by weight.

(A2-3) is composed of a propylene polymer component (B) and polypropylene-based resin (D); and as the propylene polymer component (B) and the polypropylene-based resin (D), those used in the above-mentioned (A2-1) or (A2-2) are mentioned.

When (A2-3) composed of the propylene polymer component (B) and the polypropylene-based resin (D) is used in the polypropylene-based resin composition of the present invention, preferably, the amount of the propylene-based polymer component (A1) is from 0.52 to 9.8% by weight, the amount of the propylene polymer component (B) is from 0.78 to 4.2% by weight, and the amount of the polypropylene-based resin (D) is from 98.7 to 86% by weight, wherein the total amount of the components (A1), (B) and (C) is 100 % by weight. Herein, the component (B) is used for producing a master batch of (A1).

The melt flow rate (MFR) of the polypropylene-based resin composition of the present invention is preferably from 5 to 150 g/10 minutes, more preferably from 10 to 120 g/10 minutes, from the standpoints of moldability, prevention of generation

5   of flow mark, or impact resistance.

The molecular weight distribution Q value measured by GPC of the polypropylene-based resin composition of the present invention is preferably less than 10, more preferably from 3 to 8, further preferably from 3 to 7, from the standpoint of

10   tensile elongation.

The die swell (swelling ratio) of the polypropylene-based resin composition of the present invention is preferably 1.6 or more from the standpoint of suppressing of flow mark generation on the surface of a molded article.

15   Methods of producing the polypropylene-based resin composition of the present invention is not particularly restricted, and the following methods are exemplified.

(1) A method of separately producing (A1) and (A2), and mixing them.

20   (2)A method of separately producing (A1), (B) and (C), and mixing them.

(3)A method of producing (A1) and producing a propylene-based resin (D) composed of (B) and (C) by a multi-step polymerization method, and mixing (A1) and the propylene-based resin (D).

25   (4) A method of separately producing (A1) and a part of (B), respectively, producing a propylene-based resin (D) composed of (C) and a part of (B) by a multi-step polymerization method,

and mixing (A1), a part of (B) produced separately and the propylene-based resin (D)(herein, the total amount of (B) is within the above-described range of (B) in (A2-1) or (A2-2).)

(5) A method of producing the propylene-based resin composition composed of (A1), (B) and (C) by a multi-step polymerization method.

(6) A method of producing the composition (D) composed of (C) and a part of (B) by a multi-step polymerization method, producing a master batch(E) comprising (A1) and a part of (B) by a multi-step polymerization method, and mixing (D) and (E). (herein, the total amount (B) is within the above-described range of (B) in (A2-1) or (A2-2).)

(7)A method of separately producing (A1) and a part of (B), producing a master batch(E) by mixing them, producing a propylene-based resin (D) composed of a part of (B) and (C) by a multi-step polymerization method, and mixing the master batch(E) and the propylene-based resin (D). (herein, the total amount of (B) is within the above-described range of (B) in (A2-1) or (A2-2)., and the (D) and (B) used for the master batch are the same as (2A-3) mentioned above.)

Among the above-methods, methods (5), (6) and (7) are preferable.

Production methods of the components described above are not particularly restricted, and methods of obtaining the components according to a known polymerization method using a known polymerization catalyst are listed.

As the known polymerization catalysts, for example,

catalyst systems pr pared by combining (a) a solid catalyst component containing magnesium, titanium, halogen and an electron donor as essential components, (b) an organoaluminum compound and (c) an electron donor component as a third

5 component.

As the catalyst system described above, for example, those described in U.S. Patents 4,983,561, 5,608,018 and 6,187,883 can be used. Further, with respect to polymerization methods, those described in the above-described U.S. Patents can be

10 applied.

In the method (6) or (7) described above, the method is not particularly restricted, and preferable is a method in which 98.7 to 86% by weight of the polypropylene-based resin (D) is mixed with preferably 1.3 to 14% by weight of a master batch

15 (E) comprising preferably 40 to 70% by weight, more preferably from 45 to 65% by weight of a propylene-based polymer component (A1) and preferably 60 to 30% by weight, more preferably 35 to 55 % by weight of a propylene polymer component (B)(the total amount of (A1) and (B) in the master batch(E) is 100 % by weight).

20 Regarding the compounding ratio of the polypropylene-based resin (D) to the master batch (E), more preferably, the amount of the polypropylene-based resin (D) is 98 to 88% by weight, and the amount of the propylene-based polymer composition (E) is 2 to 12% by weight. The total amount of (D) and (E) is 100 %

25 by weight.

The method of producing the master batch(E) is not particularly restricted, there are exemplified a method of

mixing above-mentioned propylene-based polymer component (A1),
and the propylene polymer component(B), and a multi-step
polymerization method of producing the (A1) in the presence
of a stereoregular polymerization catalyst system in the first
step and subsequently producing the (B) in the second or the
latter steps, or producing the (B) in the first step and
subsequently producing the (A1) in the second or the latter
steps.

More specifically, there is exemplified a method in which,
in the presence of a stereoregular polymerization catalyst
system composed of the above-mentioned solid catalyst component
(a), organoaluminum compound (b) and electron donor component
(c), at least two polymerization reactors are connected in series,
and after polymerization of the component (A1), the product
is transferred to the next polymerization vessel, and the
propylene polymerization component (B) is continuously
polymerized.

As the method of mixing the components used in the
propylene-based resin composition of the present invention,
for example, the polypropylene-based resin (D) and the master
batch (E), there are exemplified methods using kneading machines
such as a single-screw extruder, twin-screw extruder, Banbury
mixer, hot roll and the like. More specifically, the following
methods are mentioned.

(1) A method in which polymerization powder of the
polypropylene-based resin (D) and polymerization powder of the
master batch (E) are kneaded using the above-mentioned kneading

machine.

(2) A method in which polymerization powder of the polypropylene-based resin (D) and polymer powder of the master batch (E) are separately individually kneaded using a single-screw extruder or twin-screw extruder to produce pellet, thereafter, the pellet of the polypropylene-based resin (D) and the pellet of the propylene-based polymer (E) are kneaded using a single-screw extruder or twin-screw extruder.

(3) The master batch (E) previously pelletized using a single-screw extruder or twin-screw extruder is added to a kneading machine using a constant feeder and kneaded in a process of pelletizing a polymer powder of the polypropylene-based resin (D).

Preferably, in the method (2) or (3), the master batch(E) previously kneaded using a single-screw extruder or twin-screw extruder is used.

If necessary, a screen pack may be mounted on a dice of a single-screw extruder or twin-screw extruder. The screen pack to be mounted is preferably a metal fiber-sintered filter, and described, for example, in "Kikai Sekkei (Mechanical Design)" Vol. 25, No. 3, pp. 109 to 113(March,1981)".

The kneading temperature is usually from 170 to 250°C, preferably from 190 to 240°C.

In the polypropylene-based resin composition of the present invention, further 0 to 20% by weight of a propylene homopolymer (F), 10 to 35% by weight of an elastomer (G) and 2 to 30% by weight of an inorganic filler (H) can be added to

35 to 88% by weight of the polypropylene-based resin composition of the present invention, wherein the total amount of (F), (G), (H) and the polypropylene-based resin composition is 100 % by weight.

5   The propylene homopolymer (F) used in the present invention is a propylene homopolymer having an intrinsic viscosity [η] measured in tetralin at 135 °C of 2 or less obtained by homo-polymerizing propylene.

The elastomer (G) used in the present invention is not
10   particularly restricted, and preferably an elastomer containing a rubber component, and there are exemplified vinyl aromatic compound-containing rubber, ethylene-propylene random copolymer rubber, ethylene- α -olefin random copolymer rubber and elastomer composed of a mixture of them, and the
15   like.

As vinyl aromatic compound-containing rubber used in the elastomer (G), for example, a block copolymer composed of a vinyl aromatic compound polymer block and a conjugated diene-based polymer block, and the like are listed, and the
20   hydrogen addition ratio of double bonds at the conjugated diene portion is preferably 80% or more, more preferably 85% or more.

The molecular weight distribution (Q value) of the above-mentioned vinyl aromatic compound-containing rubber according to a GPC (gel permeation chromatography) method is
25   preferably 2.5 or less, more preferably 2.3 or less.

The content of vinyl aromatic compounds contained in the above-mentioned vinyl aromatic compound-containing rubber is

preferably from 10 to 20% by weight, more preferably from 12 to 19% by weight.

The melt flow rate (MFR, JIS-K-6758, 230℃) of the above-mentioned vinyl aromatic compound-containing rubber is

5 preferably from 1 to 15 g/10 minutes, more preferably from 2 to 13 g/10 minutes.

Examples of the vinyl aromatic compound-containing rubber include block copolymers such as a styrene-butadiene rubber(SBR), a styrene-butadiene-styrene rubber(SBS), a

10 styrene-isoprene-styrene rubber (SIS) and the like, and block copolymers obtained by hydrogenating these rubbers such as a styrene-ethylene-butene-styrene rubber(SEBS), styrene-ethylene-propylene-styrene rubber(SEPS) and the like. Further, rubbers obtained by reacting a vinyl aromatic compound

15 such as styrene or the like with ethylene-propylene-non-conjugated diene rubber(EPDM) are also listed. Still further, two or more vinyl aromatic compound-containing rubbers may also be used.

The method of producing the above-mentioned vinyl

20 aromatic compound-containing rubber is not particularly restricted, and for example, a method in which a vinyl aromatic compound is bonded to olefin-based copolymer rubber or conjugated diene rubber by polymerization, reaction and the like, and other methods are mentioned.

25 The ethylene-propylene random copolymer rubber used in an elastomer(G) is random copolymer rubber of ethylene and propylene, and the molecular weight distribution (Q value) of

the ethylene-propylene random copolymer rubber according to a GPC (gel permeation chromatography) method is preferably 2.7 or less, more preferably 2.5 or less.

The content of propylene contained in the above-mentioned
5   ethylene-propylene random copolymer rubber is preferably from 20 to 30% by weight, more preferably from 22 to 28% by weight.

The melt flow rate (MFR, JIS-K-6758, 190°C) of the ethylene-propylene random copolymer rubber is preferably from 1 to 15 g/10 minutes, more preferably from 2 to 13 g/10 minutes.

10   The ethylene-α-olefin random copolymer rubber used as the elastomer (G) is not particularly restricted, and random copolymer rubbers composed of ethylene and an α-olefin may be permissible. This α-olefin is an α-olefin having 4 to 12 carbon atoms, and for example, 1-butene, 1-pentene, 1-hexene,
15   1-heptene, 1-octene, 1-decene and the like are listed, and preferable are 1-butene, 1-hexene and 1-octene.

As the ethylene-α-olefin random copolymer rubber, for example, an ethylene-1-butene random copolymer rubber, ethylene-1-hexene random copolymer rubber, ethylene-1-octene
20   random copolymer rubber and the like are listed, and preferable is an ethylene-1-octene random copolymer rubber or ethylene-1-butene random copolymer rubber. Further, two or more ethylene-α-olefin random copolymer rubbers may also be used.

25   The molecular weight distribution (Q value) of the ethylene-1-octene random copolymer rubber according to a GPC (gel permeation chromatography) method is preferably 2.5 or

less, more preferably 2.3 or less.

The content of 1-octeneb unit in the ethylene-1-octene random copolymer rubber is preferably from 15 to 45% by weight, more preferably from 18 to 42% by weight.

5   The melt flow rate (MFR, JIS-K-6758, 190℃) of the above-mentioned ethylene-1-octene random copolymer rubber is preferably from 1 to 15 g/10 minutes, more preferably from 2 to 13 g/10 minutes.

The Q value of the ethylene-1-butene random copolymer 10 rubber according to a GPC method is preferably 2.7 or less, more preferably 2.5 or less.

The content of 1-butene contained in the above-mentioned ethylene-1-butene random copolymer rubber is preferably from 15 to 35% by weight, more preferably from 17 to 3% by weight.

15   The melt flow rate (MFR, JIS-K-6758, 190℃) of the above-mentioned ethylene-1-butene random copolymer rubber is preferably from 1 to 15 g/10 minutes, more preferably from 2 to 13 g/10 minutes.

The process for producing the above-mentioned 20 ethylene-propylene random copolymer rubber and ethylene-α -olefin random copolymer rubber is not particularly restricted, and they can be produced by copolymerizing ethylene and propylene, or ethylene and the α -olefin having 4 to 12 carbon atoms, using a known catalyst by a known polymerization method. As the known 25 catalyst, for example, catalyst systems composed of a vanadium compound and an organoaluminum compound and metallocene catalyst systems are listed, and as the known polymerization

method, a solution polymerization method, slurry polymerization method, high pressure ion polymerization method and gas phase polymerization method and the like are listed.

The inorganic filler (H) optionally used in the present 5 invention is not particularly restricted, and used usually for improving rigidity, and examples thereof include calcium carbonate, barium sulfate, mica, crystalline calcium silicate, talc, magnesium sulfate fiber and the like, and preferable are talc and magnesium sulfate fiber. These inorganic filler may 10 be used in combination of two or more.

Talc used as the inorganic filler (H) is not particularly restricted, and preferably one which obtained by grinding water-containing magnesium silicate. The crystal structure of the molecule of water-containing magnesium silicate is a 15 pyrophyllite type three-layer structure, and talc is formed by lamination of this structure. Talc is particularly preferably in the form of flat plate obtained by finely grinding a crystal of the molecule of water-containing magnesium silicate to about unit layer.

20 The average particle size of talc is preferably 3 μm or less. Here, the average particle size of talc means 50% equivalent particle size D_{50} obtained from an integral distribution curve of a minus mesh method measured by suspending talc in a dispersion medium such as water, alcohol or the like, 25 using a centrifugal precipitation type particle size distribution measuring apparatus.

The above-mentioned talc may be used without treatment,

alternatively, for improving dispersibility and boundary adhesion with the polypropylene-based resin composition composed of a propylene-based polymer component (A1) and a propylene-based polymer component (A2), the surface thereof

5   may be treated with known various silane coupling agents, titanium coupling agents, higher fatty acids, higher fatty esters, higher fatty amides, higher fatty salts or other surfactants.

The magnesium sulfate fiber used as the inorganic filler

10   (H) is not particularly restricted, and the average fiber length of the magnesium sulfate fiber is preferably from 5 to 50 μ m, further preferably from 10 to 30 μm. Further, the average fiber diameter of the magnesium sulfate fiber is preferably from 0.3 to 2 μm, further preferably from 0.5 to 1 μm.

15   To the polypropylene-based resin composition of the present invention, various additives may be added in an amount causing no deterioration in the object and effect thereof. Examples of the additive include antioxidants, ultraviolet absorbers, lubricants, pigments, antistatic agents, copper

20   inhibitors, flamer retardants, neutralizing agent, foaming agents, plasticizers, nucleating agents, de-foaming agents, cross-linking agents and the like. For improving heat resistance, weather resistance and anti-oxidation stability, it is preferable to added an antioxidant or ultraviolet absorber.

25   These additives, the above-mentioned elastomer (G) and the above-mentioned inorganic filler (H) may be compounded with a pellet of a polypropylene-based resin composition previously

melt-kneaded, or compounded in a stage of pelletizing a polypropylene-based resin composition.

The polypropylene-based resin composition of the present invention can be molded into an injection molded article by a known injection molding. The use of an injection molded article is particularly preferably an injection molded article for automobile, and examples thereof include a door trim, pillar, instrumental panel, bumper and the like.

EXAMPLES

The present invention is described in more detail by the following Examples and Comparative Examples below, but is not limited thereto.

The physical properties of polymers and compositions used in the Examples and Comparative Examples were measured according to the following methods.

(1) Intrinsic viscosity (unit: dl/g)

Reduced viscosities were measured at three points of concentration of 0.1, 0.2 and 0.5 g/dl, using an Ubbellohde viscometer. The intrinsic viscosity was obtained by a calculation method described in page 491 of "Kobunshi Yoeki (Polymer Solution), Kobunshi Jikkengaku 11 (1982, published by Kyoritsu Shuppan K.K.)", namely, by an extrapolation method in which reduction viscosities are plotted against concentration and the concentration is extrapolated into zero. It was measured at a temperature of 135℃, using tetralin as a solvent.

(1-1) Intrinsic viscosity of propylene-ethylene block polymer (i)

(1-1a) Intrinsic viscosity of propylene homopolymer portion (first segment of the block copolymer (i)): $[\eta]_P$

5   As sample, a propylene homopolymer was taken out of a polymerization reactor after polymerization in the first step, and the $[\eta]_P$ of the propylene homopolymer as the sample was measured.

(1-1b) Intrinsic viscosity $[\eta]_{EP}$ of propylene-ethylene
10 random copolymer portion (second segment of block copolymer (i))

Regarding the intrinsic viscosity $[\eta]_{EP}$ of a propylene-ethylene random copolymer portion, second segment of a propylene-ethylene block copolymer (i), the intrinsic
15 viscosity $[\eta]_P$ of a propylene homopolymer portion and the intrinsic viscosity $[\eta]_T$ of the whole propylene-ethylene block copolymer were measured respectively, and the weight ratio: X of a propylene-ethylene random copolymer portion to the whole propylene-ethylene block copolymer was used, and $[\eta]_{EP}$ was
20 calculated according to the following formulae (weight ratio: X to the whole propylene-ethylene block copolymer was obtained by the following measuring method (2)).

$[\eta]_{EP} = [\eta]_T/X-(1/X-1)[\eta]_P$

$[\eta]_P$: Intrinsic viscosity (dl/g) of propylene
25 homopolymer portion

$[\eta]_T$: Intrinsic viscosity (dl/g) of the whole propylene-ethylene block copolymer

(1-2) Intrinsic viscosity: $[\eta]_P$ of propylene homopolymer

The intrinsic viscosity: $[\eta]_P$ of the propylene homopolymer used in a mixture (iii) was measured according to the above-mentioned method (1).

5　(1-3) Intrinsic viscosities $[\eta]_1$ and $[\eta]_2$ of propylene-based polymer component (A1)

The intrinsic viscosity of the propylene-based polymer obtained by one-step polymerization was measured according to the above-mentioned method (1) ($[\eta]_1$). In the case of two-step

10　polymerization, the intrinsic viscosity $[\eta]_1$ of a sample taken out a polymerization reactor after the first polymerization step and the intrinsic viscosity $[\eta]_T$ of a final sample obtained after the second polymerization step were measured according to the method (1) described above. The intrinsic viscosity

15　$[\eta]_2$ of a component polymerized in the second step was calculated according to the following formulae.

$$[\eta]_2 = ([\eta]_T - [\eta]_1 \times X_1)/X_2$$

$[\eta]_T$: Intrinsic viscosity (dl/g) of the final sample after two-stage polymerization process

20　$[\eta]_1$: Intrinsic viscosity (dl/g) of the sample taken out the polymerization reactor after first polymerization step

X_1: weight ratio of component obtained in first step

X_2: weight ratio of component obtained in second step

X_1 and X_2 were respectively calculated from material

25　balance in polymerization and the total of X_1 and X_2 is 1.

The weight ratio: X of a propylene-ethylene random copolymer portion to the whole propylene-ethylene block

copolymer and the ethylene content: $(C_2')_{EP}$ of a propylene-ethylene random copolymer portion in a propylene-ethylene block copolymer were determined based on the report of Kakugo et al. (Macromolecules 1982, 15, 1150-1152)

5   from a ^{13}C-NMR spectrum measured according to the following conditions.

About 200 mg of a propylene-ethylene block copolymer was uniformly dissolved with 3 ml of orthodichlorobenzene in a 10 mmϕ test tube to prepare a sample, and the ^{13}C-NMR spectrum

10   of this sample was measured according to the following conditions.

Measuring temperature: 135℃

Pulse repeating time: 10 seconds

Pulse width: 45°

15   Accumulated frequency: 2500

(3) Isotactic pentad fraction

The isotactic pentad fraction means a fraction of propylene monomer unit existing at the center of the isotactic chain in pentad unit in a polypropylene molecule chain, in other

20   words, the chain in which five propylene monomer units are successively meso-bonded, measured by a method published in A. Zambelli et al. Macromolecules, 6, 925 (1973), namely, by using ^{13}C-NMR. The assignment of the NMR absorption peak was conducted based in Macromolecules, 8, 687 (1975) published

25   thereafter.

Specifically, the isotactic ‧ pentad fraction was measured as an area fraction of mmmm peaks in all absorption peaks in

a methyl carbon region of the ^{13}C-NMR spectrum. When the isotactic pentad of an NPL standard substance CRM No. M19-14 Polypropylene PP/MWD/2 of U.K. NATINAL PHYSICAL LABORATORY was measured by this method, it was 0.944.

5   (4) Comonomer content of propylene-based polymer component (A1)(unit: % by weight)

It was determined by measuring by an infrared spectrometry according to a method described in Polymer Handbook (1995, published by Kinokuniya Shoten), p. 616 and following pages.

10   (5) Molecular weight distribution (Q value, Mw/Mn)

It was measured by GPC (gel permeation chromatography) method under the following conditions.

Type: 150 CV type (Millipore Waters)

Column: Shodex M/S 80

15   Measuring temperature: 145℃

Solvent: orthodichlorobenzene

Sample concentration: 5 mg/8 mL

A calibration curve was made using standard polystyrene. Mw/Mn of standard polystyrene (NBS706: Mw/Mn=2.0) measured

20   under this condition was from 1.9 to 2.0.

(6) Melting peak temperature (Tm, unit: ℃)

Using a differential scanning calorimeter (DSC-7, manufactured by Perkin Elmer), a polymer was heat-treated at 220℃ for 5 minutes, then, cooled to 150℃ at a cooling rate

25   of 300℃/min. and kept at 150℃ for 1 minute, further cooled to 50℃ at a cooling rate of 5℃/min. and kept at 50℃ for 1 minute, further, heated from 50℃ to 180℃ at a heating rate

of 5℃/min, and the melting peak temperature in this operation was measured as Tm.

(7) Melt flow rate (MFR, unit: g/10 minutes)

It was measured according to JIS-K-6758. Unless otherwise stated, the measurement was carried out at a temperature of 230℃ under a load of 2.16 kg.

(8) Die swell

It was measured using Capillograph 1B manufactured by Toyo Seiki Seisakusho K.K. under the following conditions.

Measuring temperature: 220℃

L/D: 40

Shear rate: 2.432×10^3 sec^{-1}

(9) Number of granular structure (1) (unit: number/10 cm^2)

A film processed under the following conditions using a T die extruder was subjected to quantitative analysis according to the following method using an image analysis apparatus.

Film processing conditions:

A film having a width of 50 mm and a thickness of 50 μ m was produced by an extruder V-20 manufactured by Tanabe Plastic Kikai K.K. and a film drawing apparatus.

Quantitative analysis method

Images (900 dpi, 8 bit) of a film were incorporated into a computer by a scanner GT-9600 manufactured by EPSON Ltd., and the images were binarized by an image analysis soft, A zo kun manufactured by Asahi Engineering Ltd. Granular structures were recognized as lighter places than surrounding places. Since the forms of granular structures are irregular, the

diameter of a circle having the same area as that of a granular structure was recognized as the size of the granular structure, and the number of granular structures having a diameter of 200 μm or more per 10 cm^2 of a film was counted.

5   (10) Number of granular structure(2) (unit: number/43 cm^2)

        The number of granular structures of a film was counted in the same manner as in "Number of granular structure(1)" except that the area of the film was changed from 10 cm^2 to 43 cm^2.

    (11) Tensile test (UE, unit: %)

10   It was measured by a method defined in ASTM D638. Measurement was conducted using a specimen having a thickness of 3.2 mm molded by injection molding. The tensile speed was 10 mm/min, 50 mm/min or 70 mm/min, and the ultimate elongation (UE) was evaluated.

15   (12) Flexural modulus (FM, unit: kg/cm^2)

        It was measured according to a method defined in JIS-K-7203. It was measured at a measuring temperature of 23℃ at a loading speed of 2.0 mm/min or 2.5 mm/min using a specimen having a thickness of 6.4 mm and a span length of 100 mm molded by injection

20   molding.

    (13) Izod impact strength (Izod, unit: kg·cm/cm^2)

        It was measured according to a method defined in JIS-K-7110. It was measured at a measuring temperature of 23℃ or -30℃ using a notched specimen subjected to a notch processing after molding,

25   having a thickness of 6.4 mm molded by injection molding.

    (14) Heat distortion temperature (HDT, unit: ℃)

        It was measured according to a method defined in JIS-K-7207.

It was measured at a fiber stress of 18.6 kg/cm^2 or 4.6 kg/cm^2.

(15) Rockwell hardness (HR)

It was measured according to a method defined in JIS-K-7202.
It was measured using a specimen having a thickness of 3.0 mm
molded by injection molding. The measured value was indicated
in R scale.

(16) Molded article appearance (Flow mark)

A specimen of 100 mm×400 mm×3.0 mm (thickness) molded
by injection molding was used, and the appearance (flow mark)
thereof was observed visually, and judging between good and
poor was conducted.

(17) Molded article appearance (granule structure)

A specimen of 100 mm×400 mm×3.0 mm (thickness) molded
by injection molding was used, and the appearance (granule
structure) thereof was observed visually.

[Production of injection molded article]

(Production-1 of injection molded article)

The specimens (injection molded article) for evaluation
of physical properties in the above-mentioned items (11) to
(15) were prepared according to the following methods.

(1a) Specimens used in Examples 1 to 16 and Comparative Examples
1 to 13

A specimen (injection molded article) was obtained by
injection-molding a propylene-based resin composition at a
molding temperature of 220℃, a mold cooling temperature of
50℃, an injection time of 15 seconds, and a cooling time of
30 seconds, using an injection molding machine NEOMAT350/120

type manufactured by Sumitomo Heavy Industries, Ltd.

(1b) Specimens used in Examples 17 and 18 and Comparative Examples 14 and 15

5   A specimen (injection molded article) was obtained by injection-molding a propylene-based resin composition at a molding temperature of 220℃, a mold cooling temperature of 50℃, an injection time of 15 seconds, and a cooling time of 30 seconds, using an injection molding machine IS150E-V type manufactured by Toshiba Machine Co., Ltd.

10   [Production-2 of injection molded article]

The specimens for evaluation of appearance in the above-mentioned items (16) and (17) were manufactured according to the following method.

A specimen(injection molded article) was obtained by

15   injection-molding a propylene-based resin composition at a molding temperature of 220℃, a mold cooling temperature of 50℃, an injection time of 15 seconds, and a cooling time of 30 seconds, using an injection molding machine NEOMAT515/150 type manufactured by Sumitomo Heavy Industries, Ltd.

20   Synthetic methods of three kinds of catalysts (solid catalyst components (I), (II) and (III)) used for polymerizing polymers used in Examples and Comparative Examples are shown below.

(1) Solid catalyst component (I)

25   A 200 liter SUS reaction vessel equipped with a stirrer was purged with nitrogen, then, 80 liter of hexane, 6.55 mol of tetrabutoxytitanium, 2.8 mol of diisobutyl phthalate and

98.9 mol of tetraethoxysilane were charged therein to form a
uniform solution. Then, 51 liter of a 2.1 mol/L solution of
butylmagnesium chloride in diisobutyl ether was gradually added
dropwise over 5 hours while keeping the temperature in the

5   reaction vessel at 5°C. After completion of dropping, the
solution was stirred at 5°C for 1 hour and at room temperature
for 1 hour, then, the resultant was separated into solid and
liquid at room temperature, and the resulted solid was washed
with 70 L of toluene three times. Subsequently, the amount

10 of toluene was controlled so as to obtain a slurry concentration
of 0.2 kg/L, then, the slurry was stirred at 105°C for 1 hour.
Then, the slurry was cooled to 95°C, and to this was added 47.6
mol of diisobutyl phthalate, and they were reacted at 95°C for
30 minutes. After the reaction, the resultant was separated

15 into solid and liquid, and the solid was washed with toluene
twice. Subsequently, the amount of toluene was controlled so
as to obtain a slurry concentration of 0.4 kg/L, then, to this
was added 3.1 mol of diisobuty phthalate, 8.9 mol of n-dibutyl
ether and 274 mol of titanium tetrachloride, and they were

20 reacted at 105°C for 3 hours. After completion of the reaction,
the resultant was separated into solid and liquid at the same
temperature, then, thus obtained solid was washed with 90 L
of toluene twice at the same temperature. The amount of toluene
was controlled so as to obtain a slurry concentration of 0.4

25 kg/L, then, to this was added 8.9 mol of n-dibutyl ether and
137 mol of titanium tetrachloride, and they were reacted at
105°C for 1 hour. After completion of the reaction, the solution

was separated into solid and liquid at the same temperature, then, thus obtained solid was washed with 90 L of toluene three times at the same temperature, then, further washed with 70 L hexane three times, then, dried under reduced pressure, to

5   obtain 11.4 kg of a solid catalyst component. The solid catalyst component contained 1.83% by weight of a titanium atom, 8.4% by weight of a phthalate, 0.30% by weight of an ethoxy group, and 0.20% by weight of a butoxy group. Hereinafter, this solid catalyst component is referred to as solid catalyst component

10   (I).

(2) Solid catalyst component (II)

A 200 liter SUS reaction vessel equipped with a stirrer was purged with nitrogen, then, 80 liters of hexane, 6.55 mol of tetrabutoxytitanium, 2.8 mol of diisobutyl phthalate and

15   98.9 mol of tetraethoxysilane were charged therein to form a uniform solution. Then, 51 liters of a 2.1 mol/L solution of butylmagnesium chloride in diisobutyl ether was gradually added dropwise over 5 hours while keeping the temperature in the reaction vessel at 5℃. After completion of dropping, the

20   solution was stirred for 1 hour at room temperature, then, the resultant was separated into solid and liquid at room temperature, and thus obtained solid was washed with 70 L of toluene three times. Subsequently, the amount of toluene was controlled so as to obtain a slurry concentration of 0.6 kg/L, then, a mixture

25   of 8.9 mol of n-dibutyl ether and 274 mol of titanium tetrachloride was added, then, to this was further added 20.8 mol of phthalic chloride, and they were reacted at 110℃ for

3 hours. After completion of the reaction, the resulted solid was washed with toluene twice at 95℃. Then, the slurry concentration was controlled to 0.6 kg/L, then, to this was added 3.13 mol of diisobuty phthalate, 8.9 mol of n-dibutyl

5 ether and 137 mol of titanium tetrachloride, and they were reacted at 105℃ for 3 hours. After completion of the reaction, the resultant was separated into solid and liquid at the same temperature, then, the resulted solid was washed with 90 L of toluene twice at 95℃. Subsequently, the slurry concentration

10 was controlled to 0.6 kg/L, then, to this was added 8.9 mol of n-dibutyl ether and 137 mol of titanium tetrachloride, and they were reacted at 95℃ for 1 hour. After completion of the reaction, the solution was separated into solid and liquid at the same temperature, then, the resulted solid was washed with

15 90 L of toluene three times at the same temperature. Subsequently, the slurry concentration was controlled to 0.6 kg/L, then, to this was added 8.9 mol of n-dibutyl ether and 137 mol of titanium tetrachloride, and they were reacted at 95℃ for 1 hour. After completion of the reaction, the resultant

20 was separated into solid and liquid at the same temperature, then, the resulted solid was washed with 90 L of toluene three times at the same temperature, further, the solution was washed with 90 L of hexane three times, then, dried under reduced pressure, to obtain 11.0 kg of a solid catalyst component. The

25 solid catalyst component contained 1.89% by weight of a titanium atom, 20% by weight of a magnesium atom, 8.6% by weight of a phthalate, 0.05% by weight of an ethoxy group, and 0.21% by

weight of a butoxy group, and showed excellent particle properties free from fine powder. This solid catalyst component is hereinafter referred to as solid catalyst component (II).

(3) Solid catalyst component (III)

5 A 200 liter SUS reaction vessel equipped with a stirrer was purged with nitrogen, then, 80 liters of hexane, 6.55 mol of tetrabutoxytitanium, and 98.9 mol of tetraethoxysilane were charged therein to form a uniform solution. Then, 50 liters of a 2.1 mol/L solution of butylmagnesium chloride in diisobutyl

10 ether was gradually added dropwise over 4 hours while keeping the temperature in the reaction vessel at $20°C$. After completion of dropping, the solution was stirred further for 1 hour at $20°C$, then, the resultant was separated into solid and liquid at room temperature, and the resulted solid was washed with

15 70 L of toluene three times. Subsequently, toluene was taken out so as to obtain a slurry concentration of 0.4 kg/L, then, a mixture of 8.9 mol of n-dibutyl ether and 274 mol of titanium tetrachloride was added, then, to this was further added 20.8 mol of phthalic chloride, and they were reacted at $110°C$ for

20 3 hours. After completion of the reaction, the resulted solid was washed with toluene three times at $95°C$. Then, the slurry concentration was controlled to 0.4 kg/L, then, to this was added 3.13 mol of diisobuty phthalate, 8.9 mol of n-dibutyl ether and 109 mol of titanium tetrachloride, and they were

25 reacted at $105°C$ for 1 hour. After completion of the reaction, the resultant was separated into solid and liquid at the same temperature, then, washed with 90 L of toluene twice at $95°C$.

Subsequently, the slurry concentration was controlled to 0.4 kg/L, then, to this was added 8.9 mol of n-dibutyl ether and 109 mol of titanium tetrachloride, and they were reacted at 95℃ for 1 hour. After completion of the reaction, the solution

5 was separated into solid and liquid at the same temperature, then, washed with 90 L of toluene twice at the same temperature. Subsequently, the slurry concentration was controlled to 0.4 kg/L, then, to this was added 8.9 mol of n-dibutyl ether and 109 mol of titanium tetrachloride, and they were reacted at

10 95℃ for 1 hour. After completion of the reaction, the resultant was separated into solid and liquid at the same temperature, then, the resulted solid was washed with 90 L of toluene three times at the same temperature, further, the solution was washed with 90 L of hexane three times, then, dried under reduced

15 pressure, to obtain 12.8 kg of a solid catalyst component. The solid catalyst component contained 2.1% by weight of a titanium atom, 18% by weight of a magnesium atom, 60% by weight of a chlorine atom, 7.15% by weight of a phthalate, 0.05% by weight of an ethoxy group, and 0.26% by weight of a butoxy group, and

20 showed excellent particle properties free from fine powder. This solid catalyst component is hereinafter referred to as solid catalyst component (III).

[Polymerization of polymer]

(1) Production of propylene homopolymer (HPP)

25 (1-1) Production of HPP-1

(1-1a) Pre-polymerization

In a 3 liter SUS autoclave equipped with a stirrer, to

sufficiently dehydrated and deaerated hexane were added 25 mml/L of triethylaluminum (hereinafter, abbreviated as TEA), cyclohexylethyldimethoxysilane (hereinafter, abbreviated as CHEDMS) as an electron donor component at a ratio CHEDMS/TEA

5 of 0.1 (mol/mol), and 15 g/L of the solid catalyst component (I), and pre-polymerization was effected while feeding propylene continuously until the concentration thereof reached 2.5 g/g solid catalyst while keeping the temperature at 15℃ or lower. The resulted prepolymer slurry was transferred to

10 a 120 L SUS diluting vessel equipped with a stirrer and liquid butane sufficiently purified was added to this for dilution, and the solution was stored at temperatures of 10℃ or lower.

(1-1b) Main polymerization

In a fluidized bed reactor having a content volume of

15 1 m^3 equipped with a stirrer, propylene and water were fed so as to keep a polymerization temperature of 80℃, a polymerization pressure of 1.8 MPa and a hydrogen concentration in a gas phase portion of 7 vol%, and continuous gas phase polymerization was conducted while continuously feeding 62 mmol/h of TEA, 6.2 mmol/h

20 of CHEDMS and 1.2 g/h of the prepolymer slurry prepared in (1) as a solid catalyst component thereby obtaining a polymer at 20 kg/h. The resulted polymer has an intrinsic viscosity [η]$_P$ of 0.90 dl/g, and an isotactic pentad fraction of 0.97.

(1-2) Polymerization of HPP-2 to 6

25 Polymerization was conducted in the same manner as in HPP-1 except that the hydrogen concentration in a gas phase part and the feed amount of the solid catalyst component in

the main polymerization were controlled so as to obtain polymers shown in Table 1. The analysis results of the resulted polymers are shown in Table 1.

(1-3) Production of HPP-2 to HPP-6

5 (1-3a) Pre-polymerization

Polymerization was conducted in the same manner as in HPP-1 except that the solid catalyst component was the solid catalyst component(III) and the electron donor compound was t-butyl-n-propyldimethoxysilane(herein after, abbreviated as

10 tBunPrDMS)

(1-3b) Main polymerization

Main polymerization was conducted in the same manner as in HPP-1 except that the electron donor compound was tBunPrDMS, the hydrogen concentration in a gas phase part and the feeding

15 amount of a solid catalyst component in the main polymerization were controlled so as to obtain polymers shown in Table 1. The analysis results of the resulted polymers are shown in Table 1.

(2) Production of propylene-ethylene block copolymer (BCPP)

20 (2-1) Production of BCPP-1

(2-1a) Pre-polymerization

Pre-polymerization was conducted in the same manner as in HPP-1.

(2-1b) Main polymerization

25 Two fluidized bed reactors having a content volume of 1 m³ equipped with a stirrer were placed in series. a propylene polymer portion was produced in the first reactor, then,

transferred to the second reactor without deactivating the produced polymer, and a propylene-ethylene copolymer portion was continuously produced in gas phase in the second reactor. In the first reactor in the former step, propylene and hydrogen

5 were so fed as to keep a polymerization temperature of 80℃, a polymerization pressure of 1.8 MPa and a hydrogen concentration in a gas phase portion of 7.4 vol%, and under this condition, continuous gas phase polymerization was conducted while feeding 33 mmol/h of TEA, 3.3 mmol/h of CHEDMS and 0.8 g/h of the prepolymer

10 slurry prepared in (1) as a solid catalyst component thereby obtaining 18.7 kg/h of a polymer. The polymer has an intrinsic viscosity $[\eta]_p$ of 0.90 dl/g. The discharged produced polymer was fed continuously to the second reactor in the latter step. In the second reactor in the latter step, propylene, ethylene

15 and hydrogen were continuously so fed as to keep a polymerization temperature of 65℃, a polymerization pressure of 1.4 MPa, a hydrogen concentration in a gas phase portion of 4.1 vol% and an ethylene concentration of 17.2 vol%, and under this condition, continuous polymerization was continued while feeding

20 n-propymethyldimethoxysilane at 22 mmol/h, giving a polymer at 22.5 dl/g. The resulted polymer has an intrinsic viscosity $[\eta]_T$ of 1.07 dl/g, and the content of a polymer(EP content) produced in the latter step portion was 17% by weight, therefore, the polymer produced in the latter step portion (EP portion)

25 has an intrinsic viscosity $[\eta]_{EP}$ of 1.9 dl/g. The ethylene content in the EP portion was 40% by weight.

(2-2) Production of BCPP-2 and BCPP-3

Polymerization was conducted in the same manner as in BCPP-1 except that the hydrogen concentration in a gas phase part, the ethylene content and the feeding amount of a solid catalyst component in the main polymerization were controlled

5   so as to obtain polymers shown in Table 2. The analysis results of the resulted polymers are shown in Table 2.

(2-3) Production of BCPP-4 and BCPP-5

(2-3a) Pre-polymerization

Polymerization was conducted in the same manner as in HPP-1

10   except that the solid catalyst component was the solid catalyst component(III) and the electron donor compound was tBunPrDMS.

(2-3b) Main polymerization

Main polymerization was conducted in the same manner as in BPP-1 except that the electron donor compound was tBunPrDMS,

15   the hydrogen concentration in a gas phase part and the feeding amount of a solid catalyst component in the main polymerization were controlled so as to obtain polymers shown in Table 2. The analysis results of the resulted polymers are shown in Table 2.

20   (2-4)Production of BCPP-6

(2-4a) Pre-polymerization

Pre-polymerization was conducted in the same manner as in HPP-1 except that the solid catalyst component was the solid catalyst component(II) and the electron donor compound was

25   tBunPrDMS.

(2-4b) Main polymerization

Main polymerization was conducted in the same manner as in BPP-1

except that the electron donor compound was tBunPrDMS, the hydrogen concentration in a gas phase part and the feeding amount of a solid catalyst component in the main polymerization were controlled so as to obtain polymers shown in Table 2. The

5 analysis results of the resulted polymers are shown in Table 2.

[η]$_P$ shown in Table 1, and [η]$_P$, [η]$_{EP}$, ethylene content in EP and EP content shown in Table 2 are obtained by analyzing powders of propylene homopolymers (HPP-1 to HPP-6) and

10 propylene-ethylene block copolymers (BCPP-1 to BCPP-3) obtained by the above-mentioned polymerization, respectively, and MFR is a MFR of a pellet prepared by adding 0.05 parts by weight of calcium stearate (manufactured by NOF Corp.), 0.05 parts by weight of

15 3,9-bis[2-{3-(3-t-butyl-4-hydroxy-5-methylphenyl)propionyl oxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]undecan e (Sumilizer GA80, manufactured by Sumitomo Chemical Co., Ltd.) and 0.05 parts by weight of bis(2,4-di-t-butylphenyl)pentaerythritol diphosphite

20 (Ultranox U626, manufactured by GE Speciality Chemicals) as stabilizers to 100 parts by weight of a powder and granulating the mixture, using a ϕ 40 mm single-screw extruder (220℃, screen pack: 100 mesh).

(3) Production of propylene-based polymer component (A1)(HMS)

25 (3-1) Production of HMS-1

After drying under reduced pressure, into a nitrogen-purged stainless autoclave having an inner volume of

3 liter equipped with a stirr r was charged 5.1 mg of the above-mentioned solid catalyst component (II), 8.8 mmol of TEA and 88.1 μmol of di-t-butyldimethoxysilane as an electron donor component after they were allowed to contact in a heptane solution in a glass charger previously. Further, 50 mmHg of hydrogen and 780 g of propylene were charged into the above-mentioned autoclave and the mixture was heated up to 80°C to initiate polymerization. Directly after temperature rising to 80°C, an ethylene gas was continuously fed so as to give an ethylene partial pressure of 0.1 kg/m²G and polymerization was conducted for 60 minutes. 60 minutes after, the gas in the autoclave was purged, and the produced polymer was dried at 60°C for 5 hours under reduce pressure, to obtain 198.3 g of a polymer powder. The resulted powder had an intrinsic viscosity of 7.6 (dl/g) and an ethylene content of 2.7% by weight. The melting peak temperature Tm was 148.1°C. The analysis results of the resulted HMS-1 are shown in Table 3.

(3-2) Polymerization of HMS-2

Polymerization was conducted in the same manner as in HMS-1 except that the amount of the solid catalyst component (II) was changed to 5.0 mg, the hydrogen addition amount was changed to 110 mmHg and the ethylene partial pressure during polymerization was changed to 0.2 kg/m²G. The amount of the resulted polymer was 410.5 g, the intrinsic viscosity was 5.8 (dl/g) and the ethylene content was 4.5% by weight. The melting peak temperature Tm was 137.5°C. The analysis results of the resulted HMS-2 are shown in Table 3.

(3-3) Polymerization of HMS-3

(3-3a) Pre-polymerization

In a 3 liter SUS autoclave equipped with a stirrer, to sufficiently dehydrated and deaerated hexane was added TEA at

5 25 mml/L, tBunPrDMS as an electron donor component at a ratio tBunPrDMS/TEA of 0.1 (mol/mol), and the solid catalyst component (III) at 15 g/L, and pre-polymerization was effected while feeding propylene continuously until the concentration thereof reached 1 g/g solid catalyst while keeping the temperature at

10 15°C or lower. The resulted prepolymer slurry was transferred to a 120 L SUS diluting vessel equipped with a stirrer and liquid butane sufficiently purified was added to this for dilution, and the solution was stored at temperatures of 10°C or lower.

(3-3b) Main polymerization

15 In a 300 liter SUS polymerization vessel equipped with a stirrer, liquid propylene was fed at a rate of 35 kg/h so as to keep a polymerization temperature of 60°C and a slurry amount of 95 L, further, liquid 1-butene was fed so as to keep a 1-butene concentration in a gas phase portion of 15.6 vol%,

20 and continuous copolymerization of propylene-1-butene was conducted while feeding TEA at 31 mmol/h, tBunPrDMS at 4.7 mmol/h and the prepolymer slurry prepared in (3-a) as a solid catalyst component at 1.3 g/h thereby obtaining a polymer at 4.6 kg/h. The resulted polymer has an intrinsic viscosity [η] of 7.0

25 dl/g, a 1-butene content of 13.6% by weight, and a melting peak temperature of 152.5°C. The analysis results of the resulted HMS-3 are shown in Table 3.

(3-4) Production of HMS-4

(3-4a) Pre-polymerization

Pre-polymerization was conducted in the same manner as in HMS-3.

5   (3-4b) Main polymerization

In a 300 liter SUS polymerization vessel equipped with a stirrer, liquid propylene was fed at a rate of 45 kg/h so as to keep a polymerization temperature of 60℃ and a slurry amount of 95 L, further, liquid 1-butene was fed so as to keep 10   a 1-butene concentration in a gas phase portion of 5.5 vol%, and continuous copolymerization of propylene-1-butene was conducted while feeding TEA at 31 mmol/h, tBunPrDMS at 4.7 mmol/h and the previous polymer slurry prepared in (3-4a) as a solid catalyst component at 1.5 g/h thereby obtaining a polymer at 15   4.0 kg/h. The resulted polymer has an intrinsic viscosity [η] of 5.1 dl/g, a 1-butene content of 5.1% by weight, and a melting peak temperature of 154.0℃. The analysis results of the resulted HMS-3 are shown in Table 3.

(3-5) Polymerization of HMS-5

20   (3-5a) Pre-polymerization

Pre-polymerization was conducted in the same manner as in HMS-3.

(3-5b) Main polymerization

In a 300 liter SUS polymerization vessel equipped with 25   a stirrer, liquid propylene was fed at a rate of 50 kg/h so as to keep a polymerization temperature of 60℃ and a slurry amount of 95 L, and continuous polymerization of propylene was

conducted while feeding TEA at 31 mmol/h, tBunPrDMS at 4.5 mmol/h and the pre-polymer slurry prepared in (3-5a) as a solid catalyst component at 1.3 g/h, giving a polymer at 2.6 kg/h. The resulted polymer has an intrinsic viscosity [η] of 7.9 dl/g and a melting

5   peak temperature of 164.8℃. The analysis results of the resulted HMS-5 are shown in Table 3.

(3-6) Polymerization of HMS-6

(3-6a) Pre-polymerization

Pre-polymerization was conducted in the same manner as

10   for HPP-1 except that the solid catalyst component (II) was used and tBunPrDMS was used as the electron donor compound.

(3-6b) Main polymerization

In an SUS gas phase fluidized bed reactor having an inner volume of 1 m³ equipped with a stirrer, propylene, hydrogen

15   and ethylene were fed so as to keep a polymerization temperature of 60℃, a polymerization pressure of 1.8 MPa, a hydrogen concentration in a gas phase portion of 0.1 vol% and an ethylene concentration in a gas phase portion of 7.3 vol%, and continuous gas phase polymerization of propylene-ethylene was conducted

20   while feeding TEA at 60 mmol/h, tBunPrDMS at 6 mmol/h and the previous polymer slurry prepared in (3-6a) as a solid catalyst component at 0.9 g/h thereby obtaining a polymer at 22.5 kg/h. The resulted polymer has an intrinsic viscosity [η] of 5.3 dl/g, an ethylene content of 9.6% by weight and a melting peak

25   temperature of 115.6℃. The analysis results of the resulted HMS-6 are shown in Table 3.

(3-7) Production of HMS-7

(3-7a) Pre-polymerization

　　Pre-polymerization was conducted in the same manner as in HMS-3 except that the solid catalyst component (I) was used.

(3-7b) Main polymerization

5　　In a 300 liter SUS polymerization vessel equipped with a stirrer, liquid propylene was fed at a rate of 35 kg/h so as to keep a polymerization temperature of 60℃ and a slurry amount of 95 L, further, ethylene was fed so as to keep an ethylene concentration in a gas phase portion of 2.9 vol%, and continuous

10　copolymerization of propylene-ethylene was conducted in the substantial absence of hydrogen while feeding TEA at 51 mmol/h, tBunPrDMS at 5 mmol/h and the pre-polymer slurry prepared in (3-7a) as a solid catalyst component at 1.0 g/h, thereby obtaining a polymer at 6.5 kg/h. The resulted polymer was

15　transferred continuously to a second vessel without deactivation. In the second vessel, SUS fluidized bed gas phase reactor having an inner volume of 1 m^3 equipped with a stirrer, propylene and ethylene were fed so as to keep a polymerization temperature of 70℃, a polymerization pressure of 0.8 MPa and

20　an ethylene concentration in a gas phase portion of 1.8 vol%, and continuous gas phase polymerization was continued using the solid catalyst component-containing polymer transferred from the first vessel in the substantial absence of hydrogen, thereby obtaining a polymer at 16.3 kg/h. The polymer had an

25　intrinsic viscosity [η] of 8.4 dl/g, an ethylene content of 3.4% by weight and a melting peak temperature of 144.8℃. The analysis results of the resulted HMS-7 are shown in Table 3.

(3-8) Production of HMS-8

(3-8a) Pre-polymerization

Pre-polymerization was conducted in the same manner as in HMS-3 except that the solid catalyst component (I) was used.

5 (3-8b) Main polymerization

In a 300 liter SUS polymerization vessel equipped with a stirrer, liquefied propylene was fed at a rate of 35 kg/h so as to keep a polymerization temperature of 60℃ and a slurry amount of 95 L, further, liquid 1-butene was fed so as to keep

10 a 1-butene concentration in a gas phase portion of 10.1 vol%, and continuous copolymerization of propylene-1-butene was conducted in the substantial absence of hydrogen while feeding TEA at 53 mmol/h, tBunPrDMS at 4.7 mmol/h and the pre-polymer slurry prepared in (3-8a) as a solid catalyst component at 1.7

15 g/h, thereby obtaining a polymer at 4.4 kg/h. The resulted polymer has an intrinsic viscosity [η] of 8.4 dl/g, a 1-butene content of 7.4% by weight, and a melting peak temperature of 149.8℃. The analysis results of the resulted HMS-8 are shown in Table 3.

20 (3-9) Production of HMS-9

(3-9a) Pre-polymerization

Pre-polymerization was conducted in the same manner as in HMS-3 except that the solid catalyst component (I) was used.

(3-9b) Main polymerization

25 In a 300 liter SUS polymerization vessel equipped with a stirrer, liquefied propylene was fed at a rate of 35 kg/h so as to keep a polymerization temperature of 60℃ and a slurry

amount of 95 L, further, ethylene was fed so as to keep an ethylene concentration in a gas phase portion of 2.8 vol%, and continuous copolymerization of propylene-ethylene was conducted in the substantial absence of hydrogen while feeding TEA at 51 mmol/h,

5   tBunPrDMS at 5 mmol/h and the prepolymer slurry prepared in (3-9a) as a solid catalyst component at 1.0 g/h, giving a polymer at 6.1 kg/h.   The resulted polymer was transferred continuously to the second vessel without deactivation.   In the second vessel, SUS fluidized bed gas phase reaction vessel having a content

10   volume of 1 m^3 equipped with a stirrer, propylene and ethylene were fed so as to keep a polymerization temperature of 70℃, a polymerization pressure of 1.8 MPa and an ethylene concentration in a gas phase portion of 1.9 vol%, and continuous gas phase polymerization was continued using the solid catalyst

15   component-containing polymer transferred from the first vessel in the substantial absence of hydrogen, thereby obtaining a polymer at 15.7 kg/h.   The resulted polymer corresponds to a propylene-based polymer component (A1), and has an intrinsic viscosity [η] of 8.7 dl/g, an ethylene content of 3.5% by weight,

20   and a melting peak temperature of 144.8℃, and the analysis results are shown in Table 3.   Subsequently, the resulted polymer was transferred continuously to the third vessel without deactivation.   In the third vessel, SUS gas phase fluidized bed reaction vessel having an inner volume of 1 m^3 equipped

25   with a stirrer, propylene and hydrogen were fed so as to keep a polymerization temperature of 85℃, a polymerization pressure of 1.4 MPa and a hydrogen concentration in a gas phase portion

of 11.7 vol%, and continuous gas phase polymerization was continued using the polymer containing a solid catalyst component fed from the second vessel, thereby obtaining a polymer at 25.6 kg/h. The resulted polymer is HMS-9 and corresponds

5 to the propylene-based polymer composition (E), and has an intrinsic viscosity [η] of 5.7 dl/g. From the above-mentioned results, it was determined that the ratio of the total polymerization amount of the first vessel and the second vessel to the polymerization amount of the third vessel was 61:39,

10 and the polymer produced in the third vessel had an intrinsic viscosity [η] of 0.9 dl/g.

(3-10) Production of HMS-10

The polymerization was conducted in the same manner as in HMS-9 except that the ethylene concentration in a gas phase

15 part and the feeding amount of a solid catalyst component in the first vessel and second vessel and the hydrogen concentration in a gas phase part in the third vessel were changed, to obtain HMS-10 corresponding to a propylene-based polymer composition (E). The polymer produced up to the second vessel corresponds

20 to a propylene-based polymer (A1), and has an intrinsic viscosity [η] of 8.2 dl/g, an ethylene content of 2.0% by weight and a melting peak temperature of 150.8°C, and the analysis results are shown in Table 3. The ratio of the total polymerization amount of the first vessel and the second vessel to the

25 polymerization amount of the third vessel was 60:40, and the polymer produced in the third vessel had an intrinsic viscosity [η] of 1.1 dl/g.

(3-11) Production of HMS-11

The polymerization was conducted in the same manner as for HMS-10 except that ethylene fed to the first vessel and second vessel was changed to 1-butene and the 1-butene

5   concentration in a gas phase part and the feeding amount of a solid catalyst component were controlled, to obtain HMS-11 corresponding to a propylene-based polymer composition (E). The polymer produced up to the second vessel corresponds to a propylene-based polymer (A1), and has an intrinsic viscosity

10   $[\eta]$ of 7.6 dl/g, a 1-butene content of 6.5% by weight and a melting peak temperature of 150.7℃, and the analysis results are shown in Table 3. The ratio of the total polymerization amount of the first vessel and the second vessel to the polymerization amount of the third vessel was 47:53, and the

15   polymer produced in the third vessel had an intrinsic viscosity $[\eta]$ of 1.1 dl/g.

(3-12) Production of HMS-12

The polymerization was conducted in the same manner as in HMS-11 except that 1-butene concentration in a gas phase

20   portion and the feeding amount of a solid catalyst component in the first vessel and second vessel were controlled, to obtain HMS-12 corresponding to a propylene-based polymer composition (E). The polymer produced up to the second vessel corresponds to a propylene-based polymer (A1), and has an intrinsic viscosity

25   $[\eta]$ of 7.3 dl/g, a 1-butene content of 8.2% by weight and a melting peak temperature of 148.0℃, and the analysis results are shown in Table 3. The ratio of the total polymerization

amount of the first vessel and the second vessel to the polymerization amount of the third vessel was 45:55, and the polymer produced in the third vessel had an intrinsic viscosity [η] of 1.1 dl/g.

5   Example 1

0.05 parts by weight of calcium stearate (manufactured by NOF Corp.), 0.05 parts by weight of 3,9-bis[2-{3-(3-t-butyl-4-hydroxy-5-methylphenyl)propionyl oxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]undecan

10 e (Sumilizer GA80, manufactured by Sumitomo Chemical Co., Ltd.) and 0.05 parts by weight of bis(2,4-di-t-butylphenyl)pentaerythritol diphosphite (Ultranox U626, manufactured by GE Speciality Chemicals) as stabilizers were added to 100 parts by weight of a resin

15 composition composed of 20% by weight of a propylene homopolymer powder (HPP-5), 75% by weight of a propylene homopolymer powder (HPP-6) and 5% by weight of a propylene-based polymer powder (HMS-1), and they were dry-blended, then, granulated three times by a ϕ40 mm single-screw extruder (220°C, screen pack: metal

20 fiber-sintered filter NF13D manufactured by Nippon Seisen, Ltd.), to obtain a polypropylene-based resin composition. Table 4 shows compounding ratios of components, evaluation results of MFR, die swell of polypropylene-based resin compositions obtained by granulation, number-1 of granular

25 structures and mechanical properties.

Examples 2 to 4

Dry blend was effected in the same manner as in Example

1 using compounding ratios of components shown in Table 4, then, granulation was conducted to obtain polypropylene-based resin compositions. The evaluation results of MFR, die swell, number-1 of granular structures and mechanical properties are

5   shown in Table 4.

Comparative Examples 1 to 4

Dry blend was effected in the same manner as in Example 1 using compounding ratios of components shown in Table 5, then, granulation was conducted to obtain polypropylene-based resin

10   compositions. The evaluation results of MFR, die swell, number-1 of granular structures and mechanical properties are shown in Table 5.

Example 5

0.05 parts by weight of calcium stearate (manufactured

15   by NOF Corp.), 0.05 parts by weight of 3,9-bis[2-{3-(3-t-butyl-4-hydroxy-5-methylphenyl)propionyl oxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]undecan e (Sumilizer GA80, manufactured by Sumitomo Chemical Co., Ltd.) and 0.05 parts by weight of

20   bis(2,4-di-t-butylphenyl)pentaerythritol diphosphite (Ultranox U626, manufactured by GE Speciality Chemicals) as stabilizers were added to 100 parts by weight of a resin composition composed of a mixture (iii) of 65% by weight of a propylene-ethylene block copolymer (i) powder (BCPP-1) and

25   30% by weight of a propylene homopolymer powder (HPP-5), and 5% by weight of a propylene-based polymer component (MB-1), and they were dry-blended, then, granulated by a ϕ40 mm

single-screw extruder (220℃, screen pack: metal fiber-sintered filter NF13D manufactured by Nippon Seisen, Ltd.), to obtain a polypropylene-based resin composition.

5   The above-mentioned propylene-based polymer composition (MB-1) is a propylene-based polymer composition obtained by adding 0.05 parts by weight of calcium stearate (manufactured by NOF Corp.), 0.05 parts by weight of 3,9-bis[2-{3-(3-t-butyl-4-hydroxy-5-methylphenyl) propionyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro

10 [5.5]undecane (Sumilizer GA80, manufactured by Sumitomo Chemical Co., Ltd.) and 0.05 parts by weight of bis(2,4-di-t-butylphenyl) pentaerythritol diphosphite (Ultranox U626, manufactured by GE Speciality Chemicals) as stabilizers to 100 parts by weight of a resin composition

15 composed of 50% by weight of a propylene homopolymer powder (HPP-5) and 50% by weight of a propylene-based polymer powder mainly composed of propylene (HMS-7), dry-blending them, then granulation the by a ϕ 40 mm single-screw extruder (220℃, screen pack: #100 wire mesh), and corresponds to a propylene-based

20 polymer composition (E). Table 6 shows evaluation results of MFR, die swell and number-1 of granular structures of polypropylene-based resin compositions obtained by granulation at compounding ratios of components shown in Table 6.
Examples 6 to 14

25   The evaluation results of MFR, die swell and number-1 of granular structures of polypropylene-based resin compositions obtained by dry-blending in the same manner as

in Example 5 at compounding ratios of components shown in Table 6 (Examples 6 to 9) or Table 7 (Examples 10 to 14) and granulating them, are shown in Table 6 (Examples 6 to 9) or Table 7 (Examples 10 to 14), respectively.

5   (MB-2) and (MB-3) used in Examples 6 to 9 are resin compositions shown below, and correspond to a propylene-based polymer composition (E).

(MB-2): A resin composition composed of 50% by weight of a propylene homopolymer powder (HPP-5) and 50% by weight of a

10 propylene-based polymer powder (HMS-8)

(MB-3): A resin composition composed of 50% by weight of a propylene homopolymer powder (HPP-5) and 50% by weight of a propylene-based polymer powder (HMS-3)

The above-mentioned (MB-2) and (MB-3) were obtained by

15 pelletizing by the same granulation method using the same stabilizers as in (MB-1) of Example 5.

Comparative Examples 5 to 12

The evaluation results of MFR, die swell and number-1 of granular structures of polypropylene-based resin

20 compositions obtained by dry-blending in the same manner as in Example 5 at compounding ratios of components shown in Table 8 (Comparative Examples 5 to 8) or Table 9 (Comparative Examples 9 to 12) and granulating them, are shown in Table 8 (Comparative Examples 5 to 8) or Table 9 (Comparative Examples 9 to 12),

25 respectively.

(MB-4) used in Comparative Examples 5 to 8 is resin composition shown below, and correspond to a propylene-based

polymer composition (E).

(MB-4): A resin composition composed of 50% by weight of a propylene homopolymer powder (HPP-5) and 50% by weight of a propylene-based polymer powder (HMS-5)

5   The above-mentioned (MB-4) was obtained by pelletizing by the same granulation method using the same stabilizers as in (MB-1) of Example 5.

Example 15

       To 100 parts by weight of a resin composition composed
10 of 60% by weight of the polypropylene-based resin composition described in Example 1, 6% by weight of a propylene homopolymer (F) powder (HPP-4), 14% by weight of an ethylene-octene-1 random copolymer (EOR) having an octene-1 content of 24% by weight and a melt flow rate (MFR, JIS-K-6758, 190°C) of 5 g/10 minutes
15 as an elastomer (G) and 20% by weight of talc having an average particle size of 2.5 μm as an inorganic filler (H), was added the same stabilizers as used in Example 5, and the mixture was uniformly pre-mixed in a tumbler, then, the mixture was kneaded and extruded under vent suction using a twin-screw extruder
20 (TEX44SS 30BW-2V type, manufactured by The Japan Steel Works Ltd.) at an extrusion rate of 30 to 50 kg/hr and a screw revolution of 350 rpm, to produce a polypropylene-based resin composition. The screw was so constituted that a three-groove type rotor and a kneading disk were placed at the next zone to the first
25 feeding port and that to the second feeding port, namely, two positions in the kneading zone.

       Compounding ratios of components, and the evaluation

results of MFR, die swell, flow mark, granular structure appearance and mechanical properties of the resulted polypropylene-based resin composition are shown in Table 10.

Example 16

5   A propylene-based resin composition was prepared in the same manner as in Example 15 except that the polypropylene-based resin composition used in Example 15 was changed to the polypropylene-based resin composition described in Example 10.

      Compounding ratios of components, and the evaluation

10 results of MFR, die swell, flow mark, granular structure appearance and mechanical properties of the resulted polypropylene-based resin composition are shown in Table 10.

Comparative Examples 13 and 14

      A propylene-based resin composition was prepared in the

15 same manner as in Example 15 except that the polypropylene-based resin composition used in Example 15 was changed to the polypropylene-based resin composition described in Comparative Example 5 or Comparative Example 9.

      Compounding ratios of components, and the evaluation

20 results of MFR, die swell, flow mark, granular structure appearance and mechanical properties of the resulted polypropylene-based resin composition are shown in Table 10.

Example 17

      A resin composition was prepared in the same manner as

25 in Example 15 except that the polypropylene-based resin composition used in Example 15 was changed to a polypropylene-based resin composition obtained by dry-blending

49 % by weight of (BCPP-4), 45.5 % by w ight of (HPP-7) and 5.5 % by weight of (HMS-9) in the same manner as in Example 5, and then granulating the resulted mixture.

Compounding ratios of components, and the evaluation
5   results of MFR, die swell, flow mark, granular structure appearance and mechanical properties of the resulted polypropylene-based resin composition are shown in Table 11.
Example 18

A composition was prepared in the same manner as in Example
10   15 except that 100 parts by weight of the resin composition used in Example 15 was changed to 100 parts by weight of a resin composition composed of 50 parts by weight of a polypropylene-based resin composition obtained by dry-blending 53 % by weight of (BCPP-5), 43 % by weight of (HPP-8) and 4 %
15   by weight of (HMS-9) in the same manner as in Example 5 and then granulating the resulted mixture. 10 parts by weight of the propylene homopolymer (F)(HPP-4), 25 parts by weight of an ethylene-octene-1 random copolymer (EOR) having an octene-1 content of 23 % by weight and a melt flow rate(MFR, JIS-K-6758,
20   190℃) of 1g/10 min. as an elastomer (G) and 15 parts by weight of talc having an average particle size of 2.5 μm as an inorganic filler (H).

Compounding ratios of components, and the evaluation results of MFR, die swell, flow mark, granular structure
25   appearance and mechanical properties of the resulted polypropylene-based resin composition are shown in Table 11.
Example 19

A composition was prepared in the same manner as in Example

15 except that 100 parts by weight of the resin composition

used in Example 15 was changed to 100 parts by weight of a resin

composition composed of 75.5 parts by weight of a

5   polypropylene-based resin composition obtained by dry-blending

96 % by weight of (BCPP-6) and 4 % by weight of (HMS-9) in the

same manner as in Example 5 and then granulating the resulted

mixture, 8.5 parts by weight of an ethylene-octene-1 random

copolymer (EOR) having an octene-1 content of 24 % by weight

10   and a melt flow rate(MFR, JIS-K-6758, 190℃) of 5g/10 min. as

an elastomer (G) and 16 parts by weight of talc having an average

particle size of 2.5μm as an inorganic filler (H).

Compounding ratios of components, and the evaluation

results of MFR, die swell, flow mark, granular structure

15   appearance and mechanical properties of the resulted

polypropylene-based resin composition are shown in Table 11.

Table 1

		[η]P	MFR
		dl/g	g/10 minutes
Propylene homopolymer	HPP-1	0.90	140
	HPP-2	0.84	195
	HPP-3	1.48	11
	HPP-4	1.21	29
	HPP-5	0.92	120
	HPP-6	1.34	17
	HPP-7	0.95	100
	HPP-8	0.80	267

Table 2

		$[\eta]_P$	$[\eta]_{EP}$	Ethylene content in Ep	EP content	MFR
		dl/g	dl/g	wt%	wt%	g/10 min.
Ethylene-propylene block copolymer	BCPP-1	0.90	1.9	40	17	68
	BCPP-2	0.92	2.5	55	13	64
	BCPP-3	0.81	2.1	57	12	128
	BCPP-4	0.87	4.2	46	14	65
	BCPP-5	0.83	5.1	41	14	68
	BCPP-6	0.92	2.2	40	27	35

Table 3

		$[\eta]^{A1}$	Tm^{A1}
		dl/g	°C
Propylene-based polymer component (A1)	HMS-1	7.6	148.1
	HMS-2	5.8	137.5
	HMS-3	7.0	152.5
	HMS-4	5.1	154.0
	HMS-5	7.9	164.8
	HMS-6	5.3	115.6
	HMS-7	8.4	144.8
	HMS-8	8.4	149.8
	HMS-9	8.7	144.8
	HMS-10	8.2	150.8
	HMS-11	7.6	150.7
	HMS-12	7.3	148.0

65

Table 4

		Example			
		1	2	3	4
Composition (wt%)	HPP-5	20	20		
	HPP-6	75	75	95	95
	HMS-1	5			
	HMS-2		5		
	HMS-3			5	
	HMS-4				5
Mw/Mn				4.0	3.7
MFR g/10 min.		15	16	11	12
Die swell		2.14	1.92	2.04	1.79
UE(50mm/minutes) %		739	637	725	733
UE(70mm/minutes) %		108	572	769	751
FM(2.5mm/minutes) kg/cm^2		15200	14600	14700	14500
HDT(4.6kg/cm^2) ℃		118	111	117	120
IZOD23℃ kg·cm/cm		2.1	2.0	2.2	2.0
Number-1 of granular structure number/10 cm^2		3	4	0	0

Table 5

		Comparative example			
		1	2	3	4
Composition (wt%)	HPP-6	95	95	100	
	HPP-7				100
	HMS-5	5			
	HMS-6		5		
Mw/Mn		3.9	3.8	3.3	
MFR g/10 min.		10	12	17	11
Die swell		2.12	1.76	1.63	1.57
UE(50mm/minutes) %		730	631	322	766
UE(70mm/minutes) %		21	709	38	744
FM(2.5mm/minutes) kg/cm^2		15000	13700	14800	14500
HDT(4.6kg/cm^2) ℃		116	117	110	114
IZOD 23℃ kg·cm/cm		2.7	2.7	2.1	2.1
Number-1 of granular structure number/10 cm^2		0	0	-	-

Table 6

			Example				
			5	6	7	8	9
Composition (wt%)	(D)	BCPP-1	65	65			
		BCPP-2			55	55	
		BCPP-3			10	10	65
		HPP-5	30	30	30	27	
		HPP-2					27
	(E)	MB-1	5				
		MB-2		5			
		MB-3			5	8	8
MFR g/10 minutes			56	53	55	45	72
Die swell			2.13	2.09	2.05	2.22	2.28
Number-1 of granular structure number/10 cm^2			3	1	2	3	4

Table 7

			Example				
			10	11	12	13	14
Composition	(D) wt%	BCPP-1	65	65	65	65	
		BCPP-2					55
		BCPP-3					15
		HPP-5	30	30	30	30	25
		HPP-2					
	(E) wt%	HMS-9	5				5
		HMS-10		5			
		HMS-11			5		
		HMS-12				5	
MFR g/10 minutes			50	60	68	58	53
Die swell			2.15	2.17	2.13	2.11	2.21
Number-1 of granular structure number/10 cm^2			5	3	1	1	2

Table 8

			Comparative Example			
			5	6	7	8
Composition (wt%)	(D)	BCPP-1	65			
		BCPP-2		55	55	
		BCPP-3		10	10	65
		HPP-5	30	30	27	
		HPP-2				27
	(E)	MB-4	5	5	8	8
MFR	g/10 minutes		63	57	47	77
Die swell			2.08	206	2.32	2.34
Number-1 of granular structure number/10 cm^2			8	26	15	8

Table 9

			Comparative Example			
			9	10	11	12
Composition (wt%)	(D)	BCPP-1	65	65		
		BCPP-2			65	
		BCPP-3				65
		HPP-5	7			
		HPP-2	28			35
		HPP-1		35	35	
MFR	g/10 minutes		67	85	84	148
Die swell			1.39	1.40	1.37	1.36
Number-1 of granular structure number/10 cm^2			0	1	1	0

Table 10

			Example		Comparative example	
			15	16	13	14
Composition (wt%)	Resin composition	Example 5	60			
		Example 10		60		
		Comparative Example 5			60	
		Comparative Example 9				60
	(F)	HPP-4	6	6	6	6
	(G)	EOR	14	14	14	14
	(H)	talc	20	20	20	20
MFR g/10 minutes			28.4	28.2	29.8	29.3
Die swell			1.51	1.51	1.50	1.25
Flow mark (visual observation)			good	good	good	poor
Granular structure on molded article (visual observation)			◯	◯	△	◯
Number-2(>200) of granular structure number/43cm^2			0	0	4	0
YS kg/cm^2			241	244	245	238
UE(10mm/minutes) %			375	457	96	498
FM(2.0mm/minutes) kg/cm^2			22300	22700	22700	22200
IZOD 23℃ kg·cm/cm			20	19.9	17.3	19.9

IZOD -30℃	kg·cm/cm	2.5	2.4	2.4	2.5
HDT(18.6kg/cm²)	℃	75	76	76	76
HR	R scale	72	72	73	74

Granular structure on molded article (visual observation)

○: Existence of granular structure was not recognized

△: Existence of granular structure was slightly

recognized

5

Table 10

			Example		
			17	18	19
Compo- sition (wt%)	Resin composition		60	50	75.5
	(F)	HPP-4	6	10	-
	(G)	EOR	14	25	8.5
	(H)	talc	20	15	16
MFR	g/10 min.		30.0	28.7	23.0
Die swell			1.54	1.42	1.42
Flow mark (visual observation)			good	good	good
Granular structure on molded article (visual observation)			◯	◯	◯
Number-2(>200) of granular structure number/43cm^2			1	0	0
YS	kg/cm^2		257	222	173
UE(10mm/minutes) %			>500	>500	>500
FM(2.0mm/minutes) kg/cm^2			24600	17000	15200
IZOD 23℃	kg·cm/cm		19	60	63
IZOD -30℃	kg·cm/cm		2.9	5.9	5.1
HDT(18.6kg/cm^2) ℃			74	60	59
HR	R scale		81	66	44

It is understood that the compositions of Examples 1 to 19 satisfying the conditions of the present invention, when

processed into a molded article, show a low tendency of generation of flow mark, namely, high die swell, have excellent appearance such as generation of little granular structure and the like, and are excellent in balance between rigidity and

5   toughness.

On the other hand, it is understood that the composition of Comparative Example 1 shows low tensile elongation (UE) since the melting peak temperature Tm^{A1} of a propylene-based polymer component (A1) does not satisfy the requirement of the invention,

10 the composition of Comparative Example 2 shows low flexural modulus (FM) since the melting peak temperature Tm^{A1} of a propylene-based polymer component (A1) does not satisfy the requirement of the invention, and the compositions of Comparative Examples 3 and 4 show low die swell and have

15 insufficient balance between flowability (MFR) and tensile elongation (UE) since a propylene-based polymer component (A1) is not contained in the compositions.

Further, it is understood that the compositions of Comparative Examples 5 to 8 give a lot of granular structure

20 since they do not satisfy the requirement of the invention, and the compositions of Comparative Examples 9 to 12 manifest low die swell since the propylene-based polymer component (A1) is out of the scope of the invention.

It is understood that the composition of Comparative

25 Example 13 shows low tensile elongation (UE) since the melting peak temperature Tm^{A1} of a propylene-based polymer component (A1) does not satisfy the requirement of the invention, and

the composition of Comparative Example 14 shows low die swell and manifests poor flow mark appearance since the propylene-based polymer component (A1) is out of the scope of the invention.

5

As described in detail above, a polypropylene-based resin composition which, when made into a molded article, has a low tendency of generation of flow marks, namely, high die swell, has excellent appearance such as little generation of granular

10   structures and the like, and is excellent in balance of rigidity and toughness, a process for producing the same and an injection molded article made of the same can be obtained according to the present invention.